



## PATENT

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): Raymond H. Bryden

Title: SILICON CARBIDE CERAMIC COMPONENTS HAVING OXIDE LAYER

App. No.: 10/810,342

Filed:

March 26, 2004

Examiner: Brent T. O'Hern

Group Art Unit:

1772

Atty. Dkt. No.: 1035-R4303

## MS AMENDMENT

Commissioner for Patents

PO Box 1450

Alexandria, VA 22313-1450

## DECLARATION UNDER 37 C.F.R. §1.132

Sir, I hereby declare and state:

1. I received my Engineering Diploma from the University College of Cape Breton in May 1987, followed by a Bachelor's degree in Metallurgical Engineering (May 1991), Masters of Applied Science – Metallurgical (July 1993), and a PhD in Metallurgical Engineering (August 1996) all from the Technical University of Nova Scotia (Dalhousie University).

2. I have been employed by Saint-Gobain Ceramics & Plastics, Inc. since November 1996, wherein I have been mainly engaged in the research and development of advanced ceramic materials, particularly including advanced silicon carbide materials that are used for refractory applications.

3. I have reviewed the Office Action dated December 18, 2007, including the positions taken by the PTO with respect to several prior art references. I have also particularly reviewed the subject matter of US Patent 6,143,239 (Sonntag) and various secondary references, US 4,948,761 (Hida), US 4,990,469 (Dussaulx et al.) and US 4,640,889 (Hillig). For the reasons

discussed below, the prior art references, alone or in combination, fail to disclose (or suggest) all features of the claimed invention.

4. The claimed invention is drawn to a ceramic component that is composed of a ceramic body. The ceramic body comprises silicon carbide and an oxide layer. The oxide layer has particular compositional and morphological features, namely containing an amorphous phase comprising silica and a crystalline phase provided in the amorphous phase. The crystalline phase contains anisotropically-shaped crystals comprising at least one of alumina and an aluminosilicate. Of particular significance, the amorphous matrix phase and the crystalline phase making up the oxide layer form an adherent, conformal layer that resists spalling and flaking and desirably functions to passivate and protect the ceramic body from excessive oxidation. The present ceramic component may be formed through a process flow in which submicron alumina is coated onto an underlying silicon carbide substrate, followed by high temperature treatment to form the characteristic oxide layer.

5. As I understand the position taken by the PTO, the PTO has primarily relied upon Sonntag for disclosure of most of the features of the claimed invention, and on Hida for disclosure of anisotropically-shaped crystals. However, the prior art fails to disclose or suggest an oxide layer formed of an amorphous matrix phase and a crystalline phase as described above, being adherent and conformal that resists spalling and flaking and which functions to passivate and protect the ceramic body from excessive oxidation. In an effort to clearly demonstrate that the prior art fails to disclose all features of the claimed invention, I have recreated the most pertinent prior art, examples of Sonntag, and have characterized those examples.

6. The recreation of the Sonntag prior art was initiated by preparing silicon carbide substrates for sol treatment. Nitride bonded silicon carbide sampled were cut into 1 X 4 X 0.25 inch and 4 X 4 X 0.25 inch specimens, were dried in an oven at 85°C, sandblasted using silicon carbide grit to prepare and clean the surfaces for coating, and were weighed to determine untreated weight. Following substrate preparation, the substrates were coated with an AlOOH sol in accordance with the teachings of Sonntag. Particularly, a 10% solids loading AlOOH sol was obtained and stabilized with a pH adjustment. Following homogenization of the sol,

samples were placed in a large cylindrical vacuum vessel in a shallow tray containing the sol. The vacuum vessel was pumped down to 100 mBar and the samples were permitted to soak in the vacuum environment to complete infiltration. The vessel was then brought back to atmospheric pressure, and the samples were dried at 85°C. The infiltration process was done once, twice and three times for various samples.

Following sol treatment, the samples were fired at about 1450°C in a gas fired periodic kiln.

7. Following heat treatment, the samples were inspected by scanning electron microscopy (SEM). It was found that the samples created by the teachings of Sonntag had mostly a glassy outer layer with notable and extended areas of crazed or cracked areas characterized by high crystalline content. An example of the crazed areas is shown in FIG. 1 below, a characteristic of all of the samples whether infiltrated once, twice or three times. In fact, it is observed that additional infiltration exacerbated the issues with crazed areas.

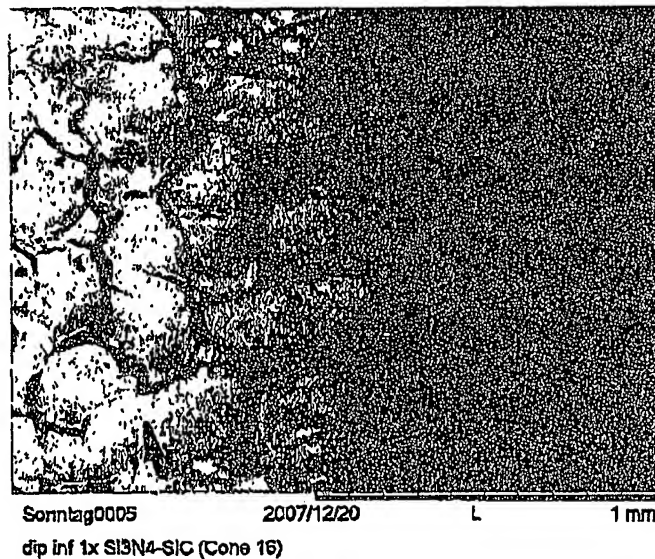
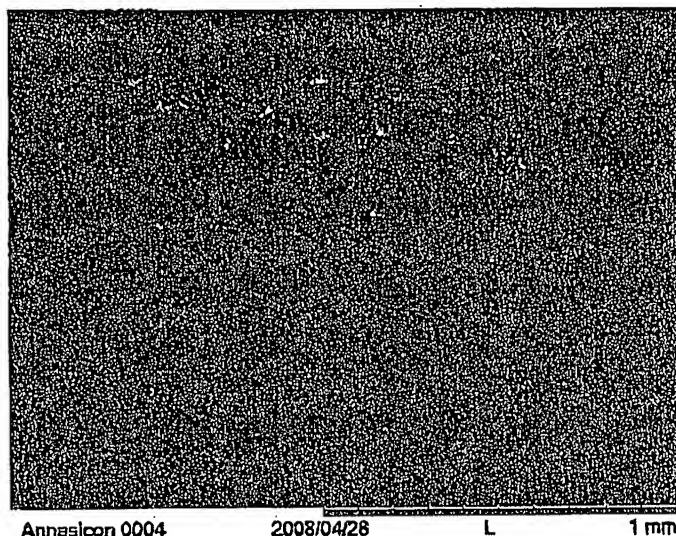


FIG. 1.

It was observed that the crazed areas were notably characterized by extended areas of large flakes, which were not adhered to the ceramic body, and easily flaked by manual scraping

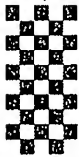
by fingernail, or pen or pencil tip. It was thus seen that the examples of Sonntag did not form an oxide layer having an amorphous matrix phase and a crystalline phase that was adherent and conformal, and which resists spalling and flaking.

8. Examples according to the claimed invention were created in a manner similar to that of the Sonntag process flow, except the slurry that was applied to the silicon carbide substrate contained  $\text{Al}_2\text{O}_3$  particles rather than  $\text{AlOOH}$  particles, and the slurry was applied manually without the complication of vacuum infiltration and without multiple applications. A representative example of those examples is shown in FIG. 2 below, showing an adherent, conformal layer that indeed notably resists spalling and flaking.



Anasicon 0004 2008/04/28 L 1 mm  
RTH  
FIG. 2.

9. In an effort to quantify the performance differences between Sonntag and that of the claimed invention, additional testing was carried out to monitor oxidation resistance, quantified in terms of weight gain. Here, samples were subjected to a steam oxidation test at  $1000^{\circ}\text{C}$  for 200 hours. The average weight gain associated with the examples of the present invention were measured to be 0.013% as compared to Sonntag examples at 0.018%. The Sonntag examples had notably inferior oxidation resistance, quantified by this 40% additional weight gain, at the



same testing conditions. It is believed that the additional weight gain characterized by the Sonntag examples is directly attributable to the non-adherent non-conformal coating, which does not effectively passivate and protect the ceramic body from oxidation.

10. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further, that these statements were made with the knowledge that willful false statements and the like, so made, are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

June 18, 2008  
Date

Raymond H. Bryden  
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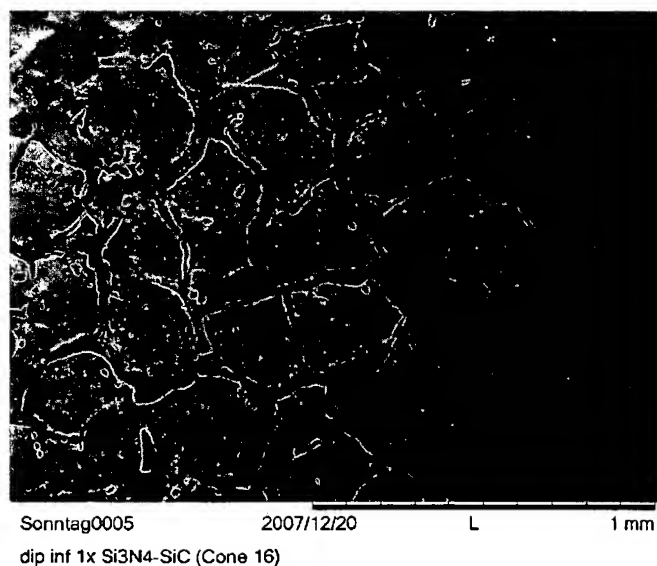


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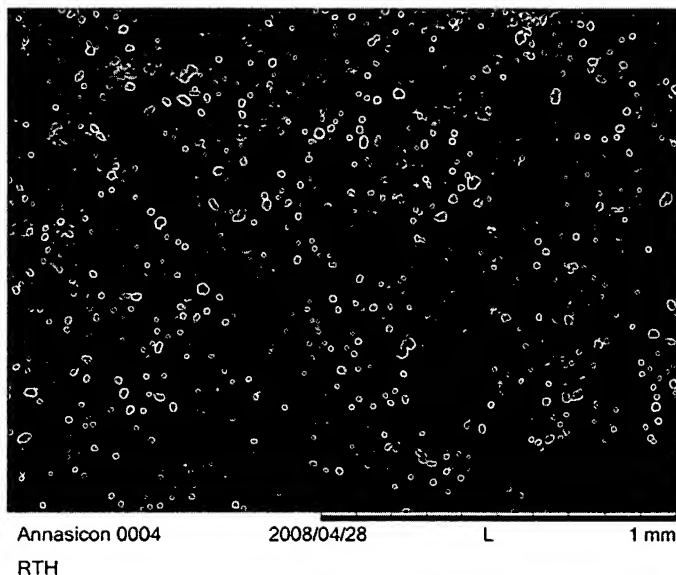


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